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Simulation of optically-stimulated luminescence of zircon

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Mineral zircon, ZrSiO_4 , belongs to a class of materials that can be used as a chronological tool in geology and archaeology for measurements of the natural radiation dose and the sample age by optical methods. The model of optically stimulated luminescence developed earlier is used to investigate the applicability of the single-aliquot regenerative-dose method (SAR) for dating with zircon. Various implementations of the SAR method are considered and practical recommendations are formulated.

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1 Introduction Natural zircon grains are subjected to internal α -, β - and γ -irradiation due to radioactive decay of U and Th impurities [1]. Typically, the contribution of external irradiation from the surrounding materials and cosmic radiation is less than 10% of total dose rate [2]. A fraction of the radiation energy is stored in the crystal in the form of electrons and holes trapped at lattice defects. The luminescent light originates from recombination of electrons and holes released from traps during thermal or optical stimulation. This effect is used to estimate the radiation dose and the time elapsed since the sample was last heated or optically bleached. Zircon has a high signal-to-noise ratio; because, unlike other minerals (such as quartz), zircon is subjected to internal irradiation at a dose rate much higher than the dose rate from external sources. Hence, it is expected that zircon is especially suitable for dating of ultra-young samples. Optically-stimulated luminescence (OSL) of zircon has not been studied systematically. Therefore, in this communication we investigate by computer simulation which OSL dating protocols should be used to minimize the errors in determination of the equivalent dose D_e .

2 The model We use the kinetic model of thermoluminescence (TL) and OSL developed in [3, 4]. Ionizing radiation creates free electron and holes, which are captured by luminescent centres (LC) and non-radiative electron or hole trapping centres. Luminescence occurs due to recombination of electrons released from traps with holes trapped by LC. The main luminescence activators in zircon are Tb^{3+} and Dy^{3+} impurity ions, which trap free holes [3, 5]. The model consists of rate equations for the concentrations of free electrons and holes, occupied electron trapping centres, occupied LC and non-radiative hole trapping centres. The assumptions and limitations of the model, the input parameters and the numerical method are discussed in detail in [3, 4]. Below we will use the set of material parameters, which has been adjusted to reproduce the observed behaviour of TL [3] and EPR [5] of irradiated zircon. In the model optical stimulation of trapped electrons to the conduction band is described by the term $\gamma_i n_i$, where γ_i is the optical excitation rate of the i -th trap and n_i is the concentration of filled light-sensitive electron traps. Following our previous paper, we will consider a special case of selective excitation of trapped electrons from the deep traps with binding energies distributed in a narrow energy interval. At $\gamma = 2 \text{ s}^{-1}$ the OSL signal decays in a few minutes. In our simulation, similar to real OSL experiments, the lumines-

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cence signal emitted by LC is filtered, i.e. we use the fraction of the OSL signal, which corresponds to the most intense Tb^{3+} band [4]. Optical dating methods are aimed at the reconstruction of the dose dependence of the luminescence response using laboratory irradiation at dose rates much higher than the natural radioactivity. In fact, the success of the optical dating depends on how close one can imitate by laboratory irradiation the natural process of trap populating. In standard optical dating the laboratory doses are administered at room temperature. However, even in the simple case of selective excitation of trapped electrons, simulation of zircon OSL reveals significant and complicated dose rate effects [4], which cannot be removed by a preheat used in dating routines. The simulation results suggest that the dose rate effects can be removed by laboratory irradiation at elevated temperatures followed by a preheat. Here we will investigate the applicability of the SAR method for dating with zircon. In this method we will consider irradiation of zircon at elevated temperatures.

2.1 The SAR protocol The SAR method has been developed for optical dating with quartz [6, 7]. Initially, the natural OSL signal L_N due to an unknown natural dose is measured from a portion of grains after a preheat. Subsequently, the same grains are used for several cycles consisting of laboratory irradiation, preheat and measurement of OSL signal L_i . The steps in each SAR cycle are simulated for zircon as follows.

Step 1 – Irradiation The simulation is started with a completely bleached sample with zero trap occupancies. This sample is “irradiated” to a natural dose $D_N = K_{nat} \times age$ at $T_{Nat}^{irr} = 20^\circ C$. The average production rate of free electrons and holes per zircon lattice site under natural conditions is $K_{nat} = 1.2 \times 10^{-17} s^{-1}$. In subsequent cycles the laboratory irradiation is simulated at elevated temperature in order (i) to prevent charge trapping by shallow traps and (ii) to produce approximately the same occupancies of the deep electron and hole traps as under natural conditions. According to [4, 9], the optimum irradiation temperature T_{lab}^{irr} depends on dose rate of laboratory irradiation K_{lab} . Here we use $T_{lab}^{irr} = 130^\circ C$ and $K_{lab} = 1.7 \times 10^7 K_{nat}$ [4].

Step 2 – Preheat 1 A preheat of 20 s at $T_{ph1} = 250^\circ C$ is applied to the sample after natural irradiation and laboratory one. Preheat 1 removes small deviations of the trap occupancies from equilibrium, i.e. it erases distinctions between natural irradiation and laboratory irradiation. According to [4] the preheat temperature should be higher than $250^\circ C$.

Step 3 – OSL measurement 1 Optical stimulation for $t_{OSL} = 150$ s at $T_{OSL} = 130^\circ C$ is performed until the intensity is nearly zero. The OSL temperature should be selected in the interval $T_{lab}^{irr} \leq T_{OSL} < T_{ph1}$. The intensity at the end of stimulation (background signal) is subtracted from the decay curve; and the resulting curve is integrated over $t_{int} \leq t_{OSL}$ to give the OSL signal $L_{N,i}$. In our simulation the accuracy of dating does not depend essentially on t_{int} .

Step 4 – Laboratory test irradiation In experiments with minerals the luminescence efficiency changes as a result of steps 1-3. To make the necessary corrections the OSL sensitivity is monitored by the OSL response L_i^t to a small constant test dose given after the main OSL measurement for each dose [7]. The value of test dose is $0.05 D_N$.

Step 5 – Preheat 2 The preheat duration is 10 s at $200^\circ C$. The purpose of this treatment is the same as for Preheat 1. To ensure that Preheat 2 does not change the occupancies of the deep traps after the first OSL measurement the temperature of Preheat 2 should be selected in the interval $T_{OSL} < T_{ph2} < T_{ph1}$.

Step 6 – OSL measurement 2 The OSL response $L_{N,i}^t$ to the test dose is measured in the same way as in OSL measurement 1.

Step 7 – Optical stimulation (bleaching) This step is not used in standard SAR. Bleaching at elevated temperature has been proposed to incorporate into the SAR protocol in order to improve the accuracy of dating [8]. In our simulation, instead of this step, for each SAR cycle we use the same initial sample identical to the sample used in the first step to simulate natural irradiation, i.e. for each SAR cycle we use a “naturally” bleached sample. To distinguish between standard SAR and SAR with additional bleaching we refer to the latter by SARB. The cycles (steps 1 to 6/7) are repeated several times for laboratory doses in the range $0.5 D_N \leq D_i \leq 1.5 D_N$. This procedure provides a regenerated growth curve of corrected

OSL response L_i/L_i^t , to which the corrected natural OSL response L_N/L_N^t should be compared to calculate the sample age.

3 Simulation results The idea of SAR and its modifications is to explore only electron traps and to get rid of the influence of LC concentrations. In the case of selective stimulation, only a small fraction of the trapped electrons is released. Large numbers of other traps compete for these electrons. For this reason the LC concentration practically does not change after OSL measurement. It can be shown that in the trap system far from saturation the integrated OSL intensity is $L \propto n_{Tb} \int_0^{t_{int}} n_e dt$, where n_{Tb} is the concentration of Tb^{4+} ions and n_e is the concentration of filled light-sensitive electron traps, which decreases to zero during stimulation (see Fig. 1). Therefore, it is expected that the corrected OSL response

does not depend on the LC concentration: $L/L^t = \int_0^{t_{int}} n_e dt \left(\int_0^{t_{int}} n_e^t dt \right)^{-1}$.

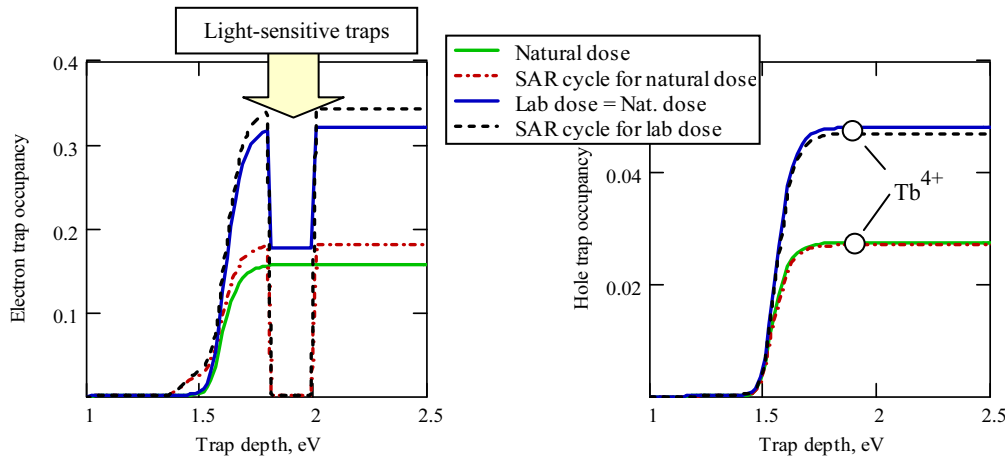


Fig. 1 The fraction of occupied electron and hole traps after the 1st SAR cycle (OSL of “natural” sample) and after the 2nd SAR cycle. The laboratory dose equals 790 Gy and corresponds to the sample age of 10^4 years. The trapped electrons are optically stimulated in the interval 1.8–2 eV.

Our attempts to find SAR parameters, which would produce acceptable accuracy, were not successful even for irradiation at optimum temperature 130 °C (Fig. 2a). The corrected OSL response is very sensitive to variation of temperature of SAR steps. The reason is the overlap of temperature effects of successive SAR steps, which influences the retrapping of electrons. Not only light-sensitive traps, but all other traps should be taken into account. During the SAR steps these traps are filled with electrons (Fig. 1).

Acceptable accuracy in dating can be obtained with the SARB protocol (Fig. 2b). In practice one can use the natural sample and several other samples which, prior to laboratory irradiation, should be reset in the same way as the natural sample (optical bleaching for sediments or annealing for pottery). Then OSL of each reset sample is measured (steps 1–3) and used to construct the dose response curve (Fig. 2b); the steps 4–6 can be skipped. We have found that SARB method is insensitive to moderate variations (± 20 °C) of the preheat temperature and the OSL temperature. Figure 2b shows that for young samples the dating accuracy depends weakly on irradiation temperature. For old samples the irradiation temperature should be close to the optimum temperature 130 °C [3, 4]. In old samples the electron and hole traps are close to saturation, therefore inaccuracies in imitation of natural irradiation affect the OSL signal.

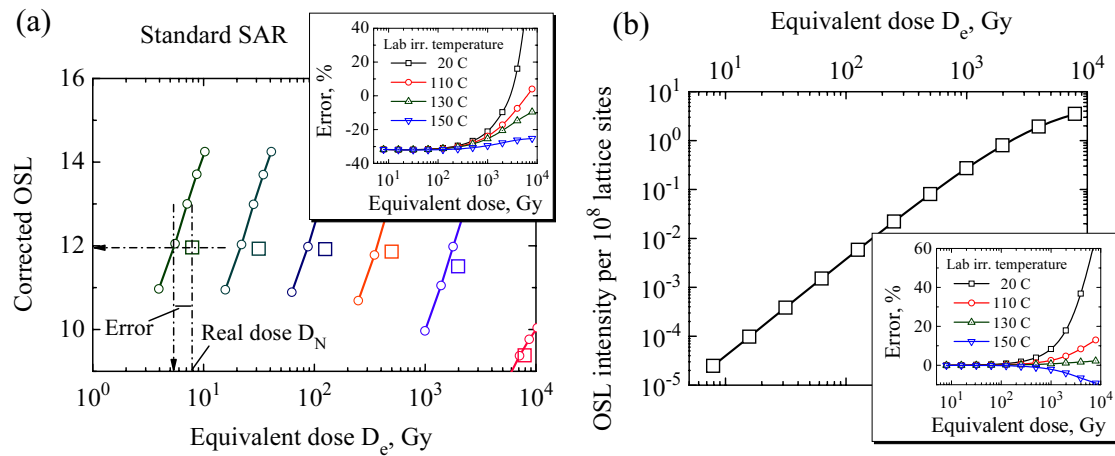


Fig. 2 Corrected OSL signal after laboratory irradiation and age determination with SAR (a), boxes correspond to natural samples. Dose dependence of OSL signal obtained with SARB (b). Inserts show the accuracy of two methods at various irradiation temperatures. The error is defined as $(D_e - D_N)/D_N$.

4 Conclusions The zircon OSL might be used successfully for dating. For the dating method to be accurate several requirements should be fulfilled:

- The initial state of the sample in each measurement cycle should be identical to the bleached natural sample. This provides a “reference state” to calibrate the natural dose using laboratory irradiation.
- To imitate natural irradiation the laboratory irradiation should be carried out at elevated temperature.
- A brief preheat before the OSL measurement diminishes dissimilarity in the occupancy of traps.
- To avoid retrapping of electrons by the shallow traps and to increase the signal, the OSL measurements should be performed at elevated temperature.

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